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Emissions of Organic Compounds and Combustion Gases during Hazardous Waste Cofiring in a Watertube Package Boiler

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EMISSIONS OF ORGANIC COMPOUNDS AND COMBUSTION GASES DURING HAZARDOUS WASTE COFIRING IN A WATERTUBE PACKAGE BOILER

by

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16. ABSTRACT

The primary objective of this study was to evaluate the sorption and desorption of organic compounds on combustion-generated soot during the cofiring of hazardous organics with fuel oil in a full-scale boiler. Sorption of organics was accomplished by firing a watertube package boiler at very low load, under a "smoking" condition, while spiking hazardous organic compounds into fuel oil. This phase was followed by a "clean" fuel firing phase using natural gas to look for any "bleed-off" of POHCs and PICs.

A secondary objective of the study was to characterize PIC emissions during the various test conditions, which is the subject of this Paper. This Paper has two major objectives: first, to evaluate and characterize PIC emissions from the combustion of specific POHCs and fuels, and second, to evaluate PIC and POHC emissions in relation to the levels of combustion gases such as carbon monoxide (CO) and to total hydrocarbons (THCs), in order to determine the usefulness of those gases as surrogates for the emissions of organic compounds.

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INTRODUCTION

EPA test results have demonstrated that industrial boilers can provide adequate thermal environments for hazardous waste destruction, achieving an average destruction and removal efficiency (DRE) of 99.998% for RCRA-listed toxic organics, i.e., principal organic hazardous constituents (POHCs) (1). Results from recent test programs, however, have suggested that unburned POHCs and products of incomplete combustion (PICs) can be adsorbed on soot deposited on boiler surfaces during cofiring and desorbed back into the combustion gases after waste cofiring ceases (2), an effect which has been termed hysteresis. Limited evidence supported this hysteresis effect, but the impact on the determination of DREs for POHCs was not known. A full-scale test was performed to determine if hysteresis actually exists and, if so, to evaluate its effect on DRE measurements. This work was performed under EPA Office of Toxic Substances (OTS) Contract No. 68-02-4252, Work Assignment 14, for the EPA Risk Reduction Engineering Laboratory.

The primary objective of this study was to evaluate the sorption and desorption of organic compounds on combustion-generated soot during the cofiring of hazardous organics with fuel oil in a full-scale boiler. Sorption of organics was accomplished by firing a watertube package boiler at very low load, under a "smoking" condition, while spiking hazardous organic compounds into fuel oil. This phase was followed by "clean" fuel firing phase using natural gas to look for any "bleed-off" of POHCs and PICs.

Preliminary findings on the subject of hysteresis have been summarized previously (3). The initial conclusions are: (1) the hysteresis effect is real and is measurable under certain conditions; (2) hysteresis can be observed both in the gas phase (stack emissions) and in the solid, sorbent phase (soot); and (3) the extent of hysteresis is compound-specific, apparently relating to physical and chemical properties such as volatility or molecular size. The impact of hysteresis upon DRE quantitation is not yet known, but it appears possible that a DRE could be affected measurably under

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some conditions. It is likely that the duration of the trial burn and the duration of the waste cofiring period which precedes it will prove to be important factors.

A secondary objective of the study was to characterize PIC emissions during the various test conditions, which is the subject of this paper. A more complete description of the overall project and findings regarding hysteresis and characterization of soot material is available (3). This paper has two major objectives: first, to evaluate and characterize PIC emissions from the combustion of specific POHCs and fuels, and second, to evaluate PIC and POHC emissions in relation to the levels of combustion gases such as carbon monoxide (CO) and to total hydrocarbons (THCs), in order to determine the usefulness of those gases as surrogates for the emissions of organic compounds.

TEST PROGRAM

Testing was conducted during September 1989 at the Tampela/Keeler (formerly Keeler/Dorr-Oliver) boiler manufacturing plant in Williamsport, Pennsylvania. The test unit was a Model DS-17.5 watertube package boiler used for plant heating and test purposes, with a capacity of 17,500 lb/h (2.21 kg/s) of steam and a furnace volume of ≈ 415 ft^3 (11.8 m^3). The boiler is fired by an air-atomizing oil burner (8:1 turndown ratio) which can accommodate several fuel types, including natural gas, No. 2 fuel oil, and No. 6 fuel oil. No air pollution control devices are installed. The unit is equipped with oxygen (O_2) and nitrogen oxide (NO_x) analyzers and an opacity meter for stack gas monitoring.

The test was designed to create conditions that were expected to allow the greatest sorption effects and also to measure those effects. During a portion of the test, operation of the boiler was deliberately altered to produce a poor combustion mode (i.e., low load and low excess air), along with nonoptimum air distribution within the burner, deliberately to produce soot.

Both monochlorobenzene (MCB) and trichloroethylene (TCE) were selected as POHCs on the basis of their thermal stability and other factors. A separate pump and metering system was set up to spike each POHC directly into the fuel oil feed line. The two POHCs and the fuel oil, which simulated a hazardous waste, were then pumped through an in-line mixer and injected into the burner. Cofiring rates ranged from about 2-1/2% to 5% for each POHC, on a volume basis.

The test program covered six different boiler operating conditions, described below. Various sampling methods were used. Soot was sampled during four conditions via specialized techniques. Stack gases were monitored for volatile organic compounds using a volatile organic sampling train (VOST) and a field gas chromatograph (GC), for semivolatile organic compounds using a Modified Method 5 (MM5) train, and for combustion gases using various continuous emission monitors (CEMs). VOST samples of 40 min duration were collected every 1 to 4 h. MM5 samples of 3- to 8-h duration were collected daily.

The first two test conditions consisted of short (4- to 6-h) background tests in which the boiler was fired with fuel only, either (1) natural gas or (2) No. 6 fuel oil. Test condition 3 used POHCs cofired with No. 6 fuel oil under nonsooting combustion conditions and involved 1 day (24 h) of continuous boiler operation.

For test condition 4, the two POHCs were cofired with fuel oil under soot-producing conditions for two continuous days. Excess air was lowered to induce soot generation. Both stack opacity readings and visual observations through boiler sight glasses confirmed soot generation. Since the test boiler was not equipped with pollution control devices, state air quality standards limited the extent of soot allowable due to a 20% stack opacity limit.

To initiate test condition 5, the fuel was switched from fuel oil to natural gas, and POHC cofiring was ceased. The combustion furnace exit temperature was maintained at the same level as in test condition 4. Under this condition the hysteresis effects of the boiler were measured for three continuous days. Test condition 6 involved a final collection of soot samples after the boiler was allowed to cool and is not discussed further in this paper.

RESULTS

Process Conditions

The boiler operating conditions and combustion gas levels for each test period are summarized in Tables 1 and 2, respectively. The combustion temperature (furnace exit temperature) was low, only ≈ 900° to 1100°F, compared with typical trial burn temperatures of 1600°F or more. Although the boiler operation is not normally controlled by furnace temperature, for this test the temperature was maintained as constant as possible.

POHC cofiring began in Run 3 at the planned rate, about 5% by volume for each POHC. The rates were reduced early into Run 4 due to extremely high levels of acid gases and organics believed to be present, and because of the potential for boiler corrosion and possible VOST analysis problems, respectively.

The stack gas flow rate ranged from 1460 to 1740 acf/min (840 to 1080 dscf/min), as measured during MM5 sampling. The stack opacity levels were generally maintained under 20% by carefully controlling excess air.

Table 2 summarizes the stack levels of gases as measured by CEMs. The oxygen (O_2) level, i.e., excess air, was reduced in Run 4 to deliberately generate soot. Since the oxygen level was manually controlled in order to maintain relatively constant stack opacity, more variation is seen in Run 4. Carbon monoxide (CO) levels were reasonably low in Runs 1, 2, and 3, but were very high in Run 4, averaging over 600 ppm. CO was initially high in Run 5, but quickly stabilized at less than 25 ppm. The total hydrocarbon (THC) trends are somewhat similar to those of CO. THC levels were very low and varied little, except during Run 4.

Table 1. Key boiler conditions.

Test condition	Approx. duration (h)	Furnace exit temp.[a] (°F)	Fuel firing rate[a]	POHC cofiring rate--volumetric basis (% total) TCE	MCB	Total	Opacity (%)
1. Natural gas (background)	4	950-1130	39 cfh (N.G.)	0	0	0	0[b]
2. Fuel oil (background)	6	965-1080	38 gph	0	0	0	0[b]
3. POHC cofiring (nonsooting)	24	935-1007	28 gph	4.5	4.4	8.9	6-8
4. POHC cofiring (sooting)	51	956-1061	28 gph	4.5/ 2.5[c]	4.6/ 2.7[c]	9.1/ 5.2[c]	11-22
5. Natural gas (hysteresis)	73	910-1070	40 cfh (N.G.)	0	0	0	0-10

Note: Nominal steam pressure was 100 psig and steam load was 2500-3000 lb/h.

[a] No. 6 fuel oil, unless noted "N.G." for natural gas.
[b] Slight baseline drift observed.
[c] Initially, cofiring rates for Run 4 continued at same rates as during Run 3. After about 3 h, cofiring was stopped for about 4 h, then resumed at about 50% of the previous rate.

Table 2. Continuous emission monitoring results.

Run No.	Average stack concentration (range shown in parentheses)[a] O_2 (%)	CO_2 (%)	CO (ppm)	THC (ppm)
1	5.0 (4.1-5.8)	8.6 (6.5-8.8)	10 (0-23)	2 (0-3)
2	5.6 (5.1-8.2)	11.5 (9.3-12.2)	64 (38-127)	2 (1-4)
3	4.8 (3.1-7.8)	12.4 (11.0-12.8)	97 (52-180)	2 (0-3)
4	2.2 (1.7-8.6)	14.5 (8.9-15.1)	626 (91-8071)	13 (1-293)
5	2.8 (0.1-11.9)	10.1 (1.4-14.9)	23 (2-828)	1 (0-26)

[a] Reported on a dry standard basis.

Stack Emissions of Organics

POHC Results. Stack gas levels of the two POHCs are shown in Figure 1 for all of the test conditions. The TCE concentration was high in the initial natural gas baseline condition (Run 1), but it decreased substantially by the end of Run 2. It is possible that the boiler was initially contaminated with TCE but the contamination was essentially "burned out" in the first few hours of operation. The background levels of MCB were relatively low.

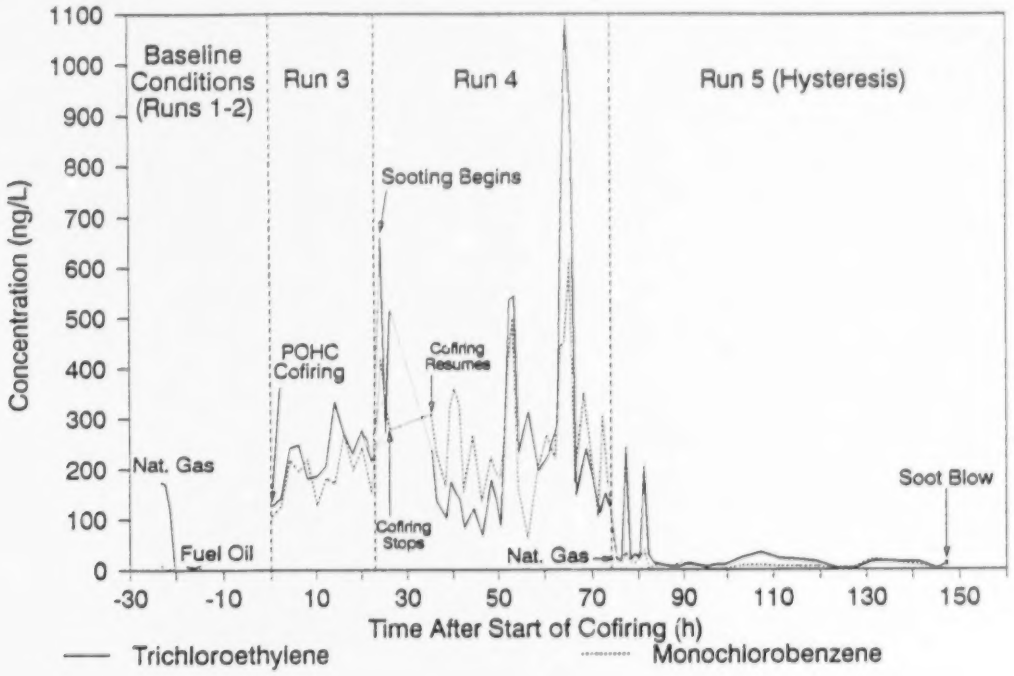

Figure 1. POHC stack gas concentrations from VOST.

When cofiring began, the POHC levels generally continued to climb during Run 3 and did not level out. The sooting condition (Run 4) began with a marked increase in stack gas POHC levels, but the levels decreased with the reduction in cofiring rate. With time, the levels generally dropped, climbed, and dropped again before the boiler feed was switched to natural gas for the hysteresis period. Some of this variation may be due to the gradual buildup of a soot layer which sorbed more and more organics from the gas stream. However, some of the more abrupt changes in concentration (i.e., "sawtooth" patterns) may be an artifact of the analysis procedures.

The POHC levels dropped rapidly upon initiation of the hysteresis condition (Run 5). Levels generally continued to drop, then stabilized

somewhat after roughly 10 h. Later in the run, the TCE levels increased, reaching a maximum roughly 33 h after the start of "hysteresis." Interestingly, the MCB levels increased and maximized even later, but were accompanied by another "peak" of TCE. At the end of the hysteresis period, soot was deliberately "blown" from the boiler bank by manually injecting steam in several pulses, to learn if POHC and PIC levels would increase. This soot blowing episode, which lasted about 10 min, resulted in a minor rise of TCE, but no MCB was detected. Some of the more volatile PICs also rose during this period.

Destruction and removal efficiencies (DREs) were determined to further evaluate combustion conditions and to better relate other test results to trial burn practices. As summarized in Table 3, DREs were generally low, only three to four "nines." DREs averaged about the same for either POHC, indicating that the actual "incinerabilities" were approximately equal under these operating conditions. (Predicted incinerabilities differ, depending upon the ranking system used.) DREs were higher in Run 3, as expected, since oxygen had not yet been lowered. DREs were higher in the first part of Run 4, compared to the second part. This was also expected, since the cofiring rate was higher, considering the generally observed trend of increasing DRE with increasing POHC concentration (4).

Table 3. DREs during POHC cofiring.

Run No.	Test condition	No. of sample pairs	Destruction/removal efficiencies (DREs): average and range (%)	
			Trichloroethylene (TCE)	Monochlorobenzene (MCB)
3	Nonsooting	11	99.9947 (99.9923-99.9975)	99.9943 (99.9913-99.9974)
4A	Sooting[a]	3	99.9904 (99.987-99.9945)	99.9913 (99.989-99.9929)
4B	Sooting[b]	27	99.986 (99.952-99.9972)	99.987 (99.970-99.9972)

[a]Initial cofiring rate nominally the same as Run 3.
[b]Adjusted cofiring rate to approximately 50% that of Runs 3 and 4A.

PIC Characterization. PICs were monitored via both volatile and semivolatile sampling techniques. VOST samples were analyzed for two POHCs and 23 other target analytes (all were EPA Method 624 compounds). The MM5 samples were analyzed for about 20 target analytes, and in addition, major unknowns were semiquantitated. The average concentrations of PICs and POHCs are compared in

Figure 2.* Total volatile PICs (i.e., all target volatile organics other than the two POHCs) were found at levels similar to the POHCs. One compound, benzene, made up much of the total PIC concentration.

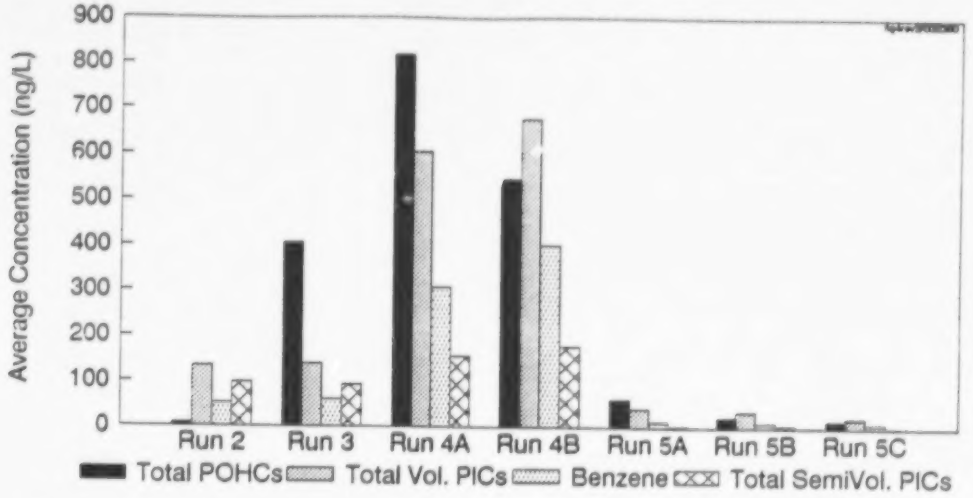

Figure 2. Average stack gas concentrations of POHCs and PICs.

The total concentration of semivolatile known PICs was less than the total (known) volatile PICs.‡ However, during Runs 2 through 4, the semivolatile unknowns were present at roughly an order of magnitude higher concentration than were the knowns. During Run 5, the semivolatile levels were quite low, and the unknowns were only about double the levels of the knowns. Over 50 unknown semivolatile compounds were found, of which 8 were present at concentrations over 100 µg/L in at least one test condition.

The average levels of the major PICs (other than benzene) are shown in Figure 3. The highest level volatile PICs included benzene, 1,1-dichloroethylene (DCE), methylene chloride ($MeCl_2$), dichlorobenzenes (DCBz's, as the total of three isomers), and toluene. The major semivolatile PICs included naphthalene and phthalic anhydride.

Levels of some of the other PICs of significance are shown in Figure 4, including the volatile compounds 1,1,1-trichloroethane (1,1,1-TCA), chloroform ($CHCl_3$), and carbon tetrachloride (CCl_4), and the semivolatile compounds phenanthrene, dibenzofuran, and benzaldehyde. Volatiles tended to predominate during Run 5. 1,1,1-TCA was found at high levels during the background runs and may have been a contaminant in the boiler.

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* Runs 4A and 4B refer to the higher and lower cofiring rate periods of the sooting condition, whereas Run 5 is divided into three segments, "A,", "B," and "C," according to when the MM5 samples were taken.

‡ The semivolatile data are blank corrected as applicable.

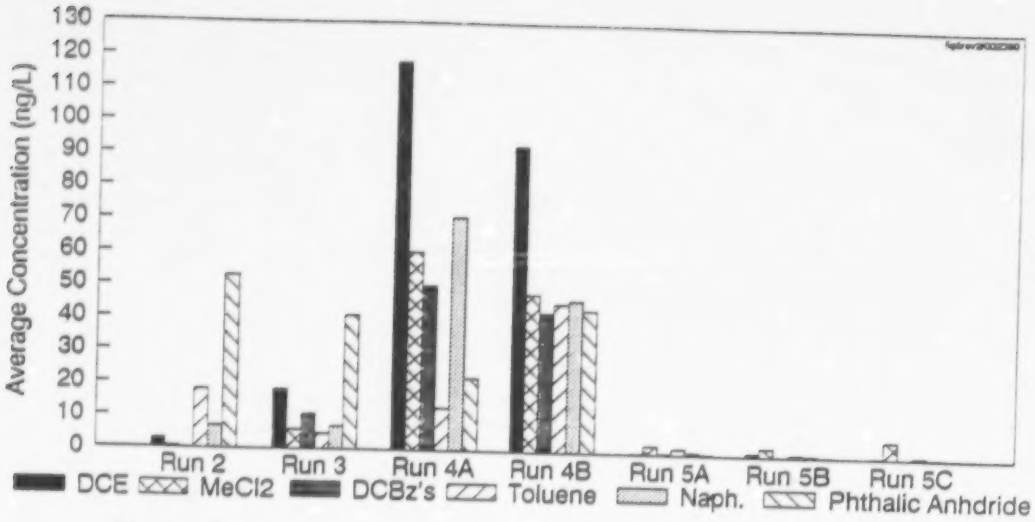


Figure 3. Average stack gas concentrations of major PICs.

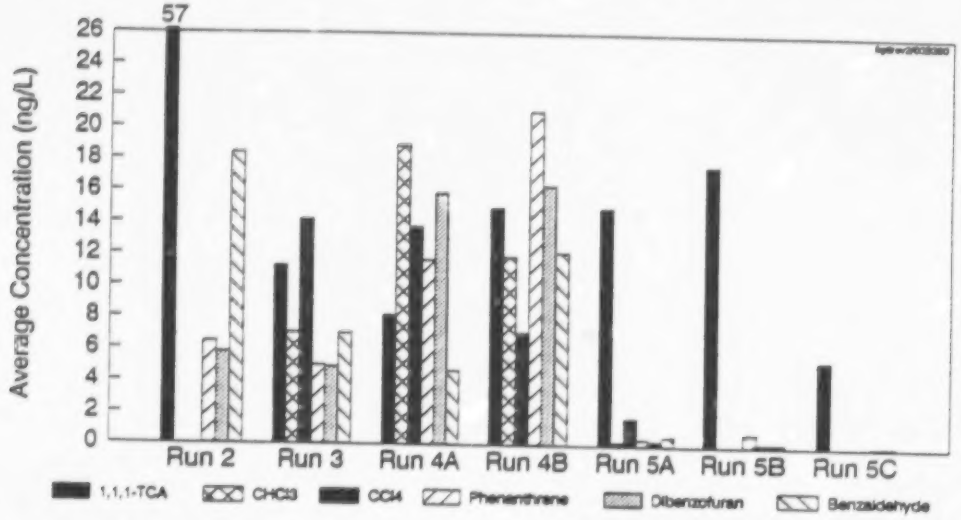


Figure 4. Average stack gas concentrations of other PICs.

Combustion Gases as Indicators of Organic Emissions

Both CO and THC have been proposed as potential surrogate indicators for organic emissions (5), with the advantage that continuous emission monitoring is possible in either case. Although some studies have shown no correlation between CO and organic emissions (6), other studies have shown that CO is a better surrogate for PICs than for POHCs, but that the relationship is not rigorous (7). Instead, CO has been envisioned as a conservative indicator of PIC emissions (i.e., high PIC levels do not necessarily accompany high CO levels, but low CO levels should ensure low PIC levels).

The current study has produced a sizable data base of POHC and PIC emissions and CO and THC levels. The levels of these constituents have varied over a wide range, and the test included very adverse combustion conditions, as evidenced by the resulting low DREs. Therefore, the relationship between the organic emissions and the combustion gases was examined for its potential value in developing an appropriate permitting approach for minimizing PIC emissions.

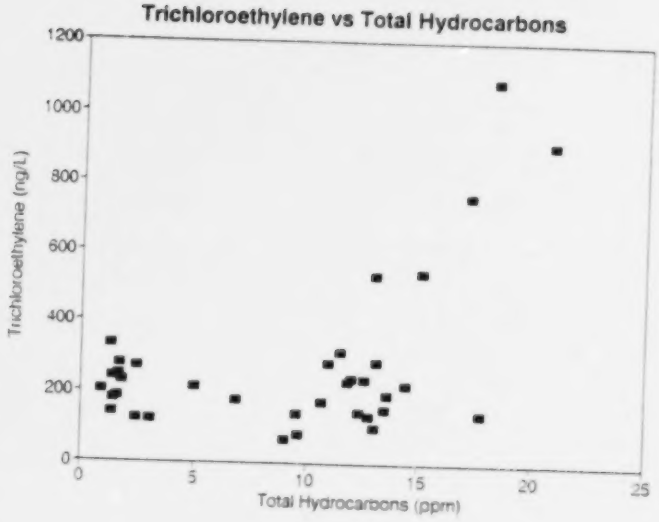
POHC and PIC emissions from this study were evaluated as a function of CO and THC levels. To accomplish this, the CEM data were divided into 40-min periods that corresponded to VOST sampling periods. Only the periods involving cofiring were evaluated (Runs 3 and 4). Using 36 "matched pairs" of data points, POHC and PIC concentrations were evaluated as a function of average CO or THC levels during the VOST sampling periods. Each POHC, total volatile PICS (target compounds only), and the major PIC (benzene) were examined via linear regression analysis.

There was no correlation (R^2=0.00) between TCE and CO and very little correlation (R^2=0.08) between MCB and CO, as expected. However, correlation between the POHCs and THC was somewhat better (R^2=0.23 for TCE and 0.33 for MCB). Scatter plots for each POHC versus THC are shown in Figures 5a and 5b.

The PICs correlated much better with the combustion gases than did the POHCs. Correlation of total volatile target PICs with CO was fair (R^2=0.42) whereas correlation with THC was good (R^2=0.81). Scatter plots for these results are shown in Figures 6a and 6b. Benzene, which was the major PIC, correlated somewhat better (R^2=0.53 for CO and 0.83 for THC). The data for benzene appear to be very similar to those for the total PICs, except that the concentrations of benzene were roughly half that of the total PICs.

Benzene has been observed as a very frequently occurring volatile PIC from hazardous waste combustion (8). It has been evaluated previously as a possible surrogate for indicating emissions of other PICs (9,10). Therefore, linear regression analysis was performed for the pairs of non-benzene volatile PICs and benzene during Runs 3 and 4. A high level of correlation (R^2=0.85) was found, as shown in Figure 7. These results suggest that benzene may have potential as a single surrogate organic compound for monitoring PICs in general. However, reliable and sensitive continuous monitoring instrumentation would also need to be developed if this approach was to be used in practice.

(a)



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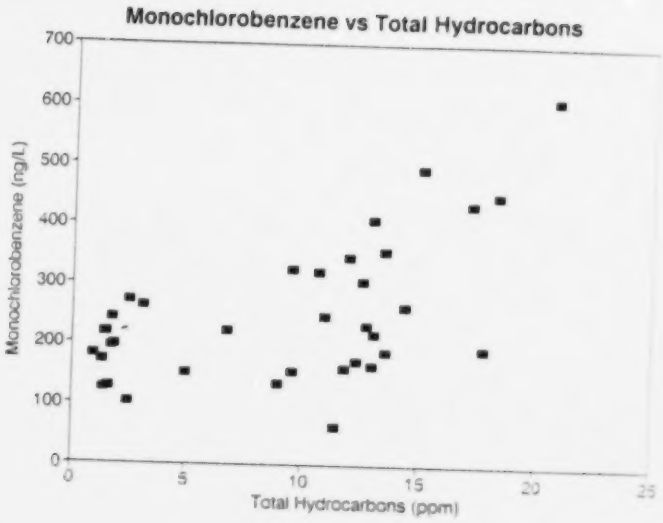
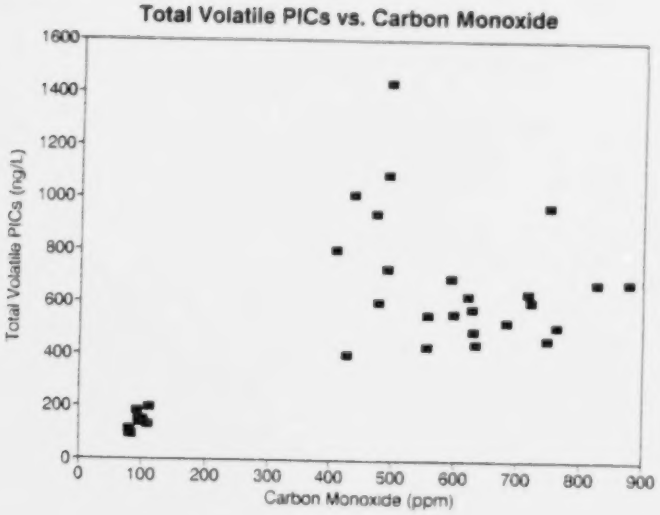


Figure 5. (a) TCE vs. THC and (b) MCB vs. THC.

(a)



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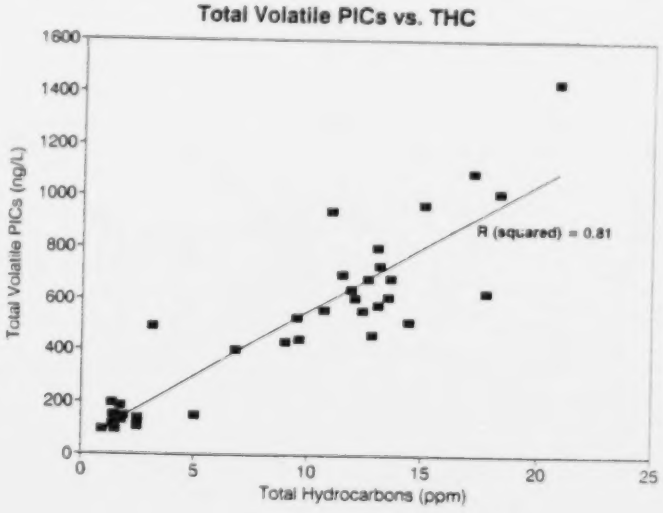

Figure 6. (a) Total volatile PICs vs. CO and (b) total volatile PICs vs. THC.

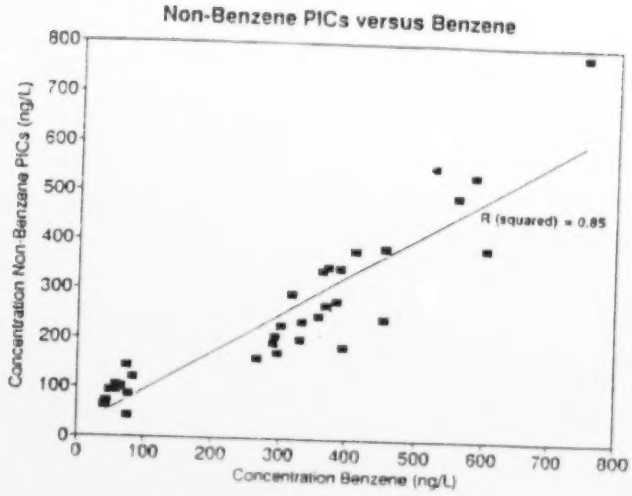

Figure 7. Non-benzene PICs vs. benzene.

CONCLUSIONS

There was little difference in DRE between TCE and MCB under both nonsooting and sooting conditions. The DREs for both POHCs fell to less than 99.99% during deliberate soot generation.

Known volatile PICs were present at higher concentration than known semivolatile PICs; however, much higher levels of unknown semivolatile PICs than knowns were observed. For the total PICs identified, total PIC concentrations were less than the total POHCs during cofiring under more favorable combustion conditions, but they exceeded the total POHCs during soot generation.

Benzene was the principal PIC identified under all conditions. The major PICs found during combustion of fuel oil only were benzene, toluene, phthalic anhydride, and benzaldehyde. During "normal" cofiring of TCE and MCB, the major PICs observed were benzene, dichloroethylene, and phthalic anhydride. During cofiring with soot generation, the major PICs observed were benzene, dichloroethylene, methylene chloride, the dichlorobenzenes, toluene, naphthalene, and phthalic anhydride.

The combustion gas measurements for CO and THC were better surrogates for PICs than for POHCs. PICs correlated better with THC than with CO. Benzene was found to correlate well with other PICs and may have potential use as a surrogate indicator for PICs in general.

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